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Interactive comment

Interactive comment on "**Argon offline-AMS source apportionment of organic aerosol over yearly cycles for an urban, rural and marine site in Northern Europe**" *by* C. Bozzetti et al.

Anonymous Referee #1

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General Comments:

This manuscript presents an analysis of the composition and source apportionment of PM1 filters collected at three sites in Lithuania. For this offline technique, the aqueous extracts from filters were nebulized with Ar for introduction into the HR-ToF-AMS. The use of Ar as the nebulization gas enabled an analysis of the CO^+/CO_2^+ fragment ratio and trends in that ratio with season. Positive matrix factorization was also applied on both the offline AMS data set as well as an offline marker data set collected using the same filters. This manuscript provides a good demonstration of the type of data sets that can be generated via this offline AMS technique and the CO^+/CO_2^+ analysis provides new insights into the interpretation of AMS data from ambient samples. Thus,

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Discussion paper

I see this paper as appropriate for publication in ACP. However, I have a few concerns, mostly related to sampling artifacts that need to be addressed prior to publication.

Specific comments:

P2 L9: Traffic exhaust OA is listed as a PMF factor from AMS spectra, yet in the experimental it is noted that the contribution is too low to be resolved with PMF and is instead estimated using a CMB approach. I suggest rewording the abstract to clarify this.

P5 L24: The nebulizer used was operated at 60 °C, how long are the aerosols in this heated region? Was this temperature in the nebulizer also used in the Daellenbach et al. analysis? What effect might this high temperature have on the composition of the organics measured with the AMS compared to online analysis? If this temperature was not used for the Daellenbach analysis, what effect might this have on the factor specific recoveries of this work compared to the results from that previous analysis?

P18 L25: PM1 composition discussed here and shown in Figure 1 shows ions that can be measured with both the AMS and IC (e.g. SO4, NO3, etc.). Do the contributions shown in Figure 1 correspond to the IC measurements or AMS? For ions that can be quantified with both techniques, how do the values compare between the AMS and IC?

P19 L14-20: The nitrate concentration shows clear seasonality with larger contributions in the winter and the sulfate concentration looks relatively constant throughout the year. However, in Figure 1, the ammonium concentration appears to also be relatively constant throughout the year. Is this correct? If so, can the authors comment on potential counter ions for NO3 ?

P20 L 28-31: The background-OOA factor appears to correlate with NH4+ much better at Preila and Vilnius than Rugsteliskes (Figure S11). Are there any potential reasons for the lower apparent correlation at Rugsteliskes? How much uncertainty is there in the NH4+ measurement? What is the significance of a correlation of B-OOA with

NH4+?

Section 2.1 and P21 L1-17: Were the High-Volume samplers located in temperature controlled rooms? If not, what effect could higher summer temperatures have on the composition of the organic compared to the winter samples? Could the S-OOA factor be complicated by collection differences caused by the loss (on the filter) of more volatile organic molecules during summer months?

Technical corrections:

P2 L6: the $CO_2^+:CO^+$ ratios reported in section 4.5 are greater than 1. The less than sign should be switched.

P10 L22-23: a verb such as "was used" is missing.

P22 L3: I suggest some mention directing the reader to Figure 5 be made in the text as the time series for the factors are discussed in this section but no mention of Figure 5 is made.

P25 L13: "Using the ratio (1.88) calculated from offline-AMS". Suggest adding OM/OCBBOA ratio to communicate what ratio is being used in the calculation here.

P30 L 25-26: suggest rephrasing, the double negative "unlikely return uncertain CO+ values" is confusing.

P45 Figure 2 and P46 Figure 4: Suggest either writing out the factor names in the labels (background-OOA instead of B-OOA etc.) or giving the names and labels in the caption.

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